Carbon monoxide pollution from cities and urban areas observed by the Terra/MOPITT mission

Cathy Clerbaux, David P. Edwards, Merritt Deeter, Louisa Emmons, Jean-François Lamarque, Xue Xi Tie, Steve T. Massie, John Gille

Carbon monoxide pollution from cities and urban areas observed by the Terra/MOPITT mission

Cathy Clerbaux,[1,2] David P. Edwards,[1] Merritt Deeter,[1] Louisa Emmons,[1] Jean-François Lamarque,[1] Xue Xi Tie,[1] Steve T. Massie,[1] and John Gille[1]

Received 21 October 2007; revised 27 November 2007; accepted 31 December 2007; published 15 February 2008.

[1] Carbon monoxide (CO) is a key species for tracking pollution plumes. The Measurement Of Pollution in The Troposphere (MOPITT) mission onboard the Terra satellite has already provided 7.5 years of CO atmospheric concentration measurements around the globe. Limited sensitivity to the boundary layer is well known to be a weakness of nadir looking thermal infrared sounders. This paper investigates the possibility of using the MOPITT surface measurements to detect CO emitted by cities and urban centers. By selecting the data and averaging them over long time periods, we demonstrate that the CO pollution arising from the large cities and urban areas can be distinguished from the background transported pollution. The more favorable observations are obtained during daytime and at locations where the thermal contrast (temperature gradient) between the surface and lower atmosphere is significant. **Citation:** Clerbaux, C., D. P. Edwards, M. Deeter, L. Emmons, J.-F. Lamarque, X. X. Tie, S. T. Massie, and J. Gille (2008), Carbon monoxide pollution from cities and urban areas observed by the Terra/MOPITT mission, *Geophys. Res. Lett.*, *35*, L03817, doi:10.1029/2007GL032300.

1. Introduction

[2] Half of the global population presently lives in urban centers. In these large urban areas air quality is becoming a major concern, and an important area of research has emerged during the last decade in order to quantify the emission sources of pollutants, to model the chemical and physical transformation that leads to the production of secondary pollutants, and to study the transport pathways for the dispersal of pollution. Carbon monoxide (CO) is a primary pollutant produced from methane and non-methane hydrocarbon oxidation, from fossil fuel combustion (associated with car traffic, industry and domestic heating) and from vegetation burning (for agricultural purposes or from wildfires). Its primary sink is oxidation by the hydroxyl radical (OH), which in turn controls the removal of most of the atmospheric pollutants as it is usually the predominant atmospheric oxidant. The CO atmospheric lifetime ranges from a few weeks to a few months depending on location and season, making it particularly suitable as a tracer of pollutant emissions.

[3] Several instruments using the Earth's thermal emission, the reflected solar radiation, or solar occultation have demonstrated that CO tropospheric concentrations can be measured from space [e.g., *Clerbaux et al.*, 2002; *Deeter et al.*, 2003; *Frankenberg et al.*, 2005; *McMillan et al.*, 2005; *Clerbaux et al.*, 2005; *Edwards et al.*, 2006a; *Buchwitz et al.*, 2007; *Luo et al.*, 2007]. The MOPITT (Measurement Of Pollution in The Troposphere) instrument [*Drummond and Mand*, 1996], launched in 1999 onboard the Terra satellite, has been providing nearly continuous measurements of CO since March 2000. MOPITT views the Earth over all latitudes with a pixel size of 22 km by 22 km and a cross-track swath that measures a near-global distribution of CO every 3 days. This extended observation period has led to a better understanding of the CO global scale variability [*Edwards et al.*, 2004; *Clerbaux et al.*, 2004; *Edwards et al.*, 2006b], to the improved quantification of regional CO emission sources [*Arellano et al.*, 2004; *Pétron et al.*, 2004; *Yudin et al.*, 2004; *Stavrakou and Müller*, 2006; *Lin et al.*, 2007], to the identification of the amount of CO released during fire events [*Lamarque et al.*, 2003; *Pfister et al.*, 2005; *Turquety et al.*, 2007] and to the study of convective transport [*Kar et al.*, 2004]. In a recent paper, *Deeter et al.* [2007] used comparisons of simulated weighting functions and averaging kernels with operational MOPITT data to show that the MOPITT sensitivity to lower tropospheric CO varies substantially depending on surface-lower atmosphere thermal contrast. This paper extends that work, and for the first time demonstrates the MOPITT potential to detect enhanced levels of CO at city scale using multi-year observations. We discuss the MOPITT ability to detect CO over urban and industrialized areas using measurements at the surface level, and show the instrument sensitivity in the lowermost atmosphere under specific conditions.

2. MOPITT Measurements and Detection of Local Pollution Plumes

2.1. MOPITT CO Measurements

[4] The MOPITT remote sensor was designed to operate by sensing infrared (IR) radiation from either thermal IR emission/absorption at 4.7 μm for CO profiles, or reflected solar radiation in the near-IR at 2.2–2.4 μm for CO and methane column measurements during daytime. The measurement technique exploits gas correlation radiometry to determine tropospheric concentrations. Due to instrumental issues with the near-IR channels, the retrieved CO profiles are currently based exclusively on thermal-IR channel radiances [*Deeter et al.*, 2003]. The operational MOPITT retrieval algorithm is based on the Optimal Estimation

[1] Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, Colorado, USA.
[2] Now at UPMC Univ. Paris 6, CNRS UMR 7620, Service d'Aéronomie/IPSL, Paris, France.

0094-8276/08/2007GL032300

Method [*Rodgers*, 2000] and delivers CO mixing ratios at 7 altitudes. The number of independent pieces of vertical information was assessed to range between 0.5 and 2 (where less than 1 means that a-priori information is a major component of the retrieved product, and larger than 1 indicates some profile shape information) [*Deeter et al.*, 2004]. The Level 2 (L2) pixel measurement products are provided with a full characterization in terms of associated error covariance matrices, allowing the calculation of the averaging kernel functions and the measurement errors required to exploit the data along with other measurements or model calculations. These retrievals have been validated with aircraft in-situ measurements [*Emmons et al.*, 2004]. For this paper we used the L2 Version 3 (v3) retrieved products.

2.2. Detection of CO Plumes

[5] With its relatively long lifetime, CO can be transported trans-boundary, and most nations are simultaneously sources of pollution and recipients of another country's pollution. CO loading is generally higher in the Northern Hemisphere than in the Southern Hemisphere, reflecting greater anthropogenic activity and associated sources. During the summer months, CO levels are lower in the Northern Hemisphere than during winter, as solar radiation results in high OH levels and more active photochemistry, and thus plumes are easier to detect above the reduced background burden. Due to the occurrence of boreal fires and to the large emissions associated with coal burning practices and transportation [e.g., *Streets et al.*, 2003] persistent high CO values are observed above China. In the Southern Hemisphere, recurrent vegetation burning during the dry season over Africa and South America generates high CO concentrations. The CO plumes emitted locally spread from regional to global scales.

[6] MOPITT was not designed to detect local pollution plumes emitted from cities. The large pixel size (22 km $\times$ 22 km) and the long revisit time interval are drawbacks to use these datasets for operational air quality purposes. Moreover, over the oceans, as emphasized by *Deeter et al.* [2004], the measurement has its maximum sensitivity to CO in the lower free troposphere and lacks sensitivity to the planetary boundary layer (PBL) due to reduced thermal contrast (the gradient in temperature between the surface and the first atmospheric layers). Over land, it was already shown that in the vicinity of Mexico City elevated mixing ratios are observed [*Massie et al.*, 2006], and that some enhanced sensitivity to the CO in the lower troposphere is to be expected when the thermal contrast is important [*Deeter et al.*, 2007].

[7] Our study uses the full 7.5 years of available MOPITT global data. The main goal is the identification of CO pollution plumes arising from cities and urban areas. We also examine the conditions under which these essentially continuous emissions can be distinguished from the background burden. The latter is an important consideration in the case of CO where zonal concentrations can be high due to the medium CO lifetime and show significant seasonal and interannual variability [*Edwards et al.*, 2004]. In order to separate the continuous localized signal generated by urban activity from the changing background, we have averaged the MOPITT L2 data over long time periods. This increases the precision due to the redundancy of the accumulated information. Figures 1 and 2 illustrate the average of the CO mixing ratios at the surface level, as derived from the measurements from March 2000 to June 2007. Only cloud-free daytime measurements, and observations corresponding to an a-priori contribution lower than 50%, were used in this calculation. Because we focused on daytime observations over land, where the highest thermal contrast is expected [*Deeter et al.*, 2007], we achieve maximum information content along with an increased sensitivity towards the surface.

[8] Figure 1 compares the global L2 MOPITT surface CO distribution to population density [CIESIN, 2000, http://sedac.ciesin.columbia.edu/gpw], both averaged over a $1^\circ \times 1^\circ$ grid, over China and parts of India and Japan, which are among the most populated areas of the globe. The EDGAR CO emission inventory [*Olivier et al.*, 1999] (excluding biomass burning) for the same part of the globe is also shown. A very good correlation, especially for China, is observed between the MOPITT surface CO mixing ratios and the population density, with the highest CO levels observed where the higher population density occurs. In China and India, the combination of old car fleet and large use of coal as anthropogenic fuels contributes to elevated levels of CO. As an example, the use of coal during 2003 amounted to 1502 Tg in China and 976 Tg in the US, respectively [*Tie et al.*, 2006]. Figure 1 (bottom) shows that the systematic use of satellite data to constrain the emission sources might help for locations where one lacks measurements. In contrast, over Europe and the US, the level of CO is lower and the signal is a continuous mixing of the background CO loading (long-range transported from China and the USA, and from the boreal fires in Siberia and Canada) and the local production of urban areas.

[9] Figure 2 presents 2000–2007 time averages of MOPITT CO over specific regions (US west and east coasts, the Milan area (Italy), Mexico City, Teheran (Iran), Tokyo (Japan), Moscow (Russia), Jakarta (Indonesia) and Johannesburg (South Africa)), where intense emissions originating from large cities can be observed. The enhancement of CO can clearly be detected, either above the city, or in the general vicinity, such as the Pô valley in Italy where pollution accumulates south of the Alps. It should be noted that for some of these cities we found a surprisingly few number of observational data points. This might be due either to high aerosols loading above cities, and subsequent flagging of these data as cloud-contaminated, or a specific problem in the V3 treatment that filters out some high CO values.

[10] The most favorable situations to observe these plumes are 1) places surrounded by mountains such as Milan, Jakarta, Teheran and Mexico City, where the pollution is trapped and the source is isolated to some extent from the surroundings, and 2) locations where high thermal contrast conditions are found, such as Tokyo, Moscow, LA-San Diego, San Francisco, NY-Philadelphia and Johannesburg. For the latter case, the high CO may also be related to the large power facilities as identified in satellite retrievals of NO_2 from *Boersma et al.* [2007]. It is worth mentioning that some notably polluted cities in Africa (e.g., Lagos,

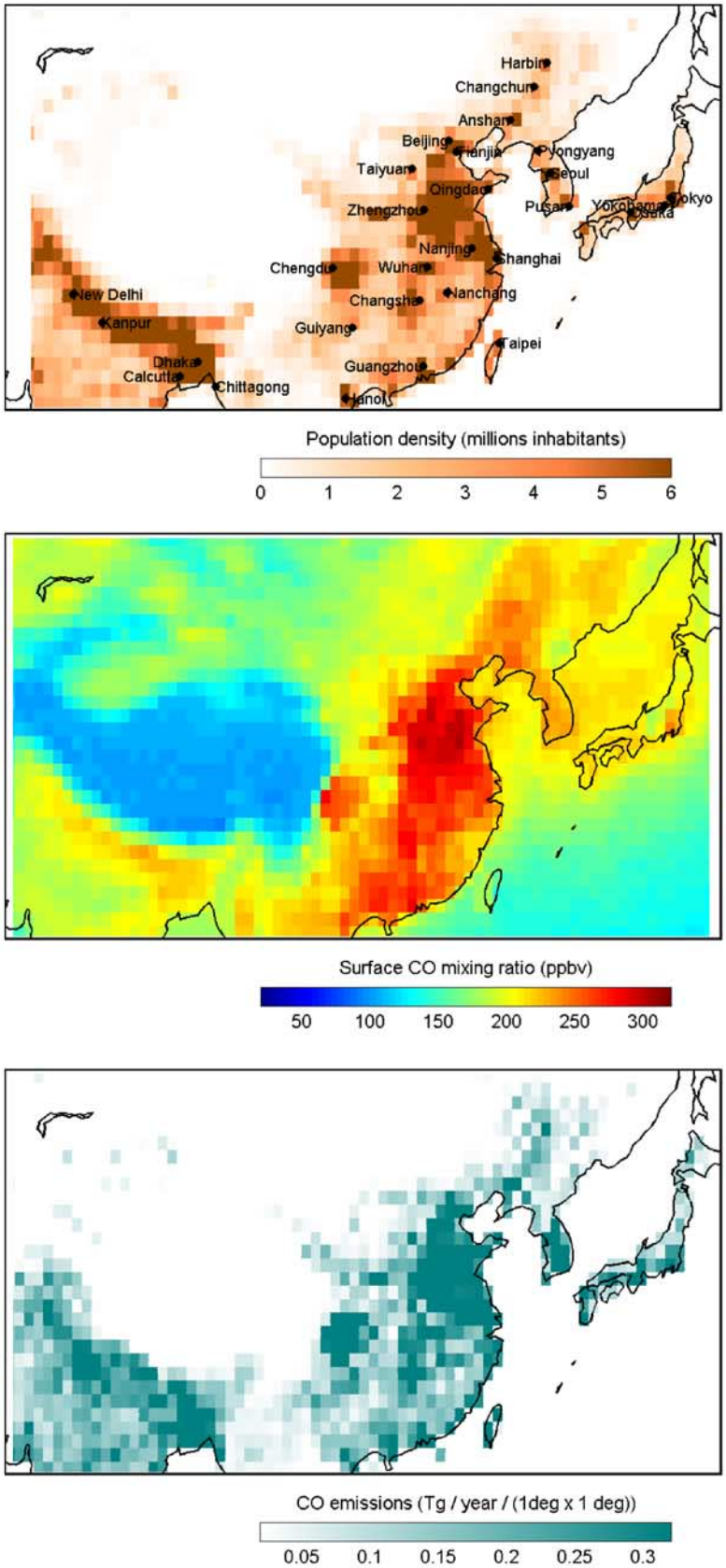

Figure 1. (top) Population density (source CIESIN, in million inhabitants) over China and surroundings. All cities with more than 2 millions of inhabitants are indicated. (middle) MOPITT CO mixing ratios at the surface level (obtained by averaging the MOPITT L2 measurements from March 2000 to June 2007). (bottom) The EDGAR CO emission inventory for year 2000. All the data are on a $1^\circ \times 1^\circ$ grid.

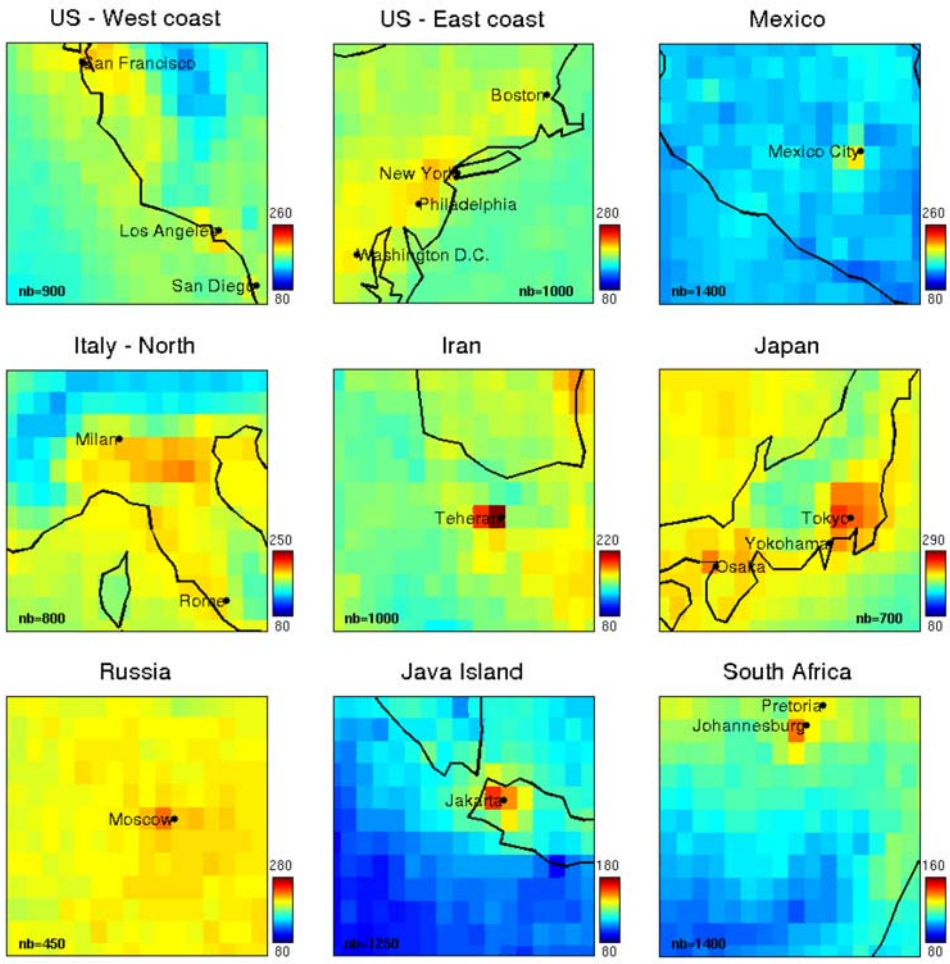

Figure 2. MOPITT CO mixing ratios at the surface level (obtained by averaging the MOPITT L2 measurements from March 2000 to June 2007, on a $0.5^\circ \times 0.5^\circ$ grid) over selected places. The linear color bars were scaled to each maximum for sake of clarity, with the lower limit fixed at 80 ppbv. The averaged number of data at the city location is indicated at the bottom of each subplot.

Nigeria) and South America (e.g., Sao Paulo, Brazil) cannot be detected, firstly as they are often under the outflow coming from regional biomass burning activity, and secondly because thermal contrast conditions are relatively weak, presumably due to evaporation and evapotranspiration processes associated with surrounding forests [*Deeter et al.*, 2007].

[11] An analysis of the averaging kernel information content at the location of several cities was performed following the methodology described by *Deeter et al.* [2007], using data properly normalized to remove vertical grid effects. Figure 3 provides an illustration of the grid-normalized averaging kernels for four MOPITT scenes over 2004 (single measurements, and averaged). As can be seen, over Johannesburg and Teheran, the averaging kernel associated with the CO surface data peaks low in the atmosphere most of the time, with a maximum just above the surface. Conversely, over Lagos and Sao Paulo the averaging kernel associated with the surface data is most influenced by CO around 600 hPa, with weak or no information coming from the surface. The observation is therefore sensitive to CO from emission sources that have risen out of the PBL and have been diluted.

3. Conclusion and Discussion

[12] Given the paucity of ground-based measurements of ambient air pollution for some cities, the possibility of using satellite-derived observations to monitor pollution sources and transport should be investigated. Satellites can sample the whole planet and could complement the in-situ ground stations network for air quality forecasts. Unlike NO_2, which is a good tracer of urban pollution due to its short lifetime [*Wang et al.*, 2007], the medium lifetime of CO makes it difficult to distinguish from space different source types and the relative contributions of local production and transported pollution.

[13] This work demonstrates that infrared thermal instruments such as MOPITT are able to detect the CO signature coming from urban areas, although the signal is limited by the low thermal contrast, the high CO global burden in some cases, and the current instrumental specifications. The more favorable MOPITT observations of cities or urban spots are

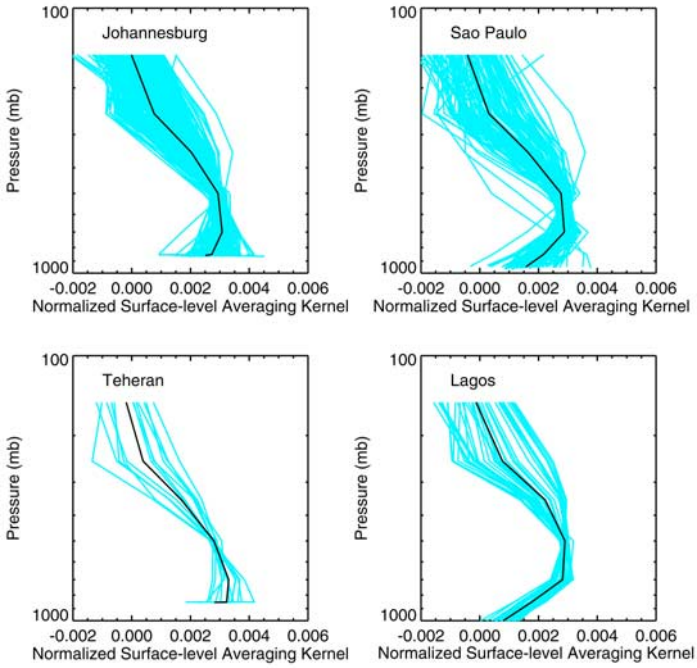

Figure 3. Grid-normalized surface level averaging kernels above Johannesburg (South Africa), Teheran (Iran), Sao Paulo (Brazil) and Lagos (Nigeria) as observed by MOPITT in 2004. The blue lines correspond to single observations and the black line is an average over all observations.

obtained at cloud-free locations where the temperature gradient (thermal contrast) between the surface and lower atmosphere is significant (daytime, over land and at places not surrounded by vegetation). Although the CO signature arising from cities might to some extent be amplified by the specific dynamic and thermal conditions occurring over urban areas (heat island effect), a detailed analysis of all locations with more than 4 million inhabitants showed that both conditions (high CO and good thermal contrast) need to be fulfilled in order to identify pollution at the city scale using the MOPITT observations. Using the whole 7 year record, only a few cities around the world can be detected, with the notable exception of China for which a strong correlation between population density and CO emission is observed all over the country.

[14] It is worth noting that MOPITT was designed to sound atmospheric CO using both thermal infrared and solar radiation. The current MOPITT CO products were retrieved using the thermal infrared channels only, but work is currently ongoing at NCAR to also exploit the solar channels. Recent papers using MOPITT and SCIAMACHY measurements [*Buchwitz et al.*, 2007; *Turquety et al.*, 2008] have shown that the combined use of these two complementary observations led to improved information in the boundary layer.

[15] New space-borne missions are currently being designed for air quality measurements in support of policy decisions. Future instruments aiming at pollution tracking should have higher revisit times (using geo-stationary or drifting orbits), with smaller and agile footprints (to maximize the occurrence of cloud-free pixels), and should sound deeper in the atmosphere (by improving instrumental specifications in terms of signal to noise ratios and spectral resolution). Last but not least, different types of sensors (using multi-spectral approaches and/or different sounding geometry) should fly onboard the same satellite.

[16] **Acknowledgments.** We acknowledge the reviewers for their useful comments, and C. Granier, J. Hadji-Lazaro and S. Turquety (Service d'Aéronomie) for their contributions. This work was undertaken in the framework of the European Network of Excellence on Atmospheric Composition Change (ACCENT-AT2). The gridded population density data were obtained from the Center for International Earth Science Information Network (CIESIN), Columbia University; and Centro Internacional de Agricultura Tropical, available at http://sedac.ciesin.columbia.edu/gpw. We wish to thank the MOPITT team at NCAR, Boulder, Colorado, USA, for data processing and algorithm support, and at the University of Toronto, Ontario, Canada, for instrument operations. The MOPITT team at NCAR acknowledges NASA's EOS Program (grants NNG04GA45G, NNG06GB27G and NNG04GO58G). The National Center for Atmospheric Research is sponsored by the National Science Foundation.

References

Arellano, A. F., P. S. Kasibhatla, L. Giglio, G. R. van der Werf, and J. T. Randerson (2004), Top-down estimates of global CO sources using MOPITT measurements, *Geophys. Res. Lett.*, *31*(1), L01104, doi:10.1029/2003GL018609.

Boersma, K. F., et al. (2007), Near-real time retrieval of tropospheric NO2 from OMI, *Atmos. Chem. Phys.*, *7*, 2103–2118.

Buchwitz, M., I. Khlystova, H. Bovensmann, and J. P. Burrows (2007), Three years of global carbon monoxide from SCIAMACHY: Comparison with MOPITT and first results related to the detection of enhanced CO over cities, *Atmos. Chem. Phys.*, *7*, 2399–2411.

Clerbaux, C., et al. (2002), Retrieval of CO from nadir remote-sensing measurements in the infrared by use of four different inversion algorithms, *Appl. Opt.*, *41*(33), 7068–7078.

Clerbaux, C., J. Gille, and D. Edwards (2004), New directions: Infrared measurements of atmospheric pollution from space, *Atmos. Environ.*, *38*, 4599–4601.

Clerbaux, C., P.-F. Coheur, D. Hurtmans, B. Barret, M. Carleer, R. Colin, K. Semeniuk, J. C. McConnell, C. Boone, and P. Bernath (2005), Carbon monoxide distribution from the ACE-FTS solar occultation measurements, *Geophys. Res. Lett.*, *32*, L16S01, doi:10.1029/2005GL022394.

Deeter, M. N., et al. (2003), Operational carbon monoxide retrieval algorithm and selected results for the MOPITT instrument, *J. Geophys. Res.*, *108*(D14), 4399,doi:10.1029/2002JD003186.

Deeter, M. N., L. K. Emmons, D. P. Edwards, J. C. Gille, and J. R. Drummond (2004), Vertical resolution and information content of CO profiles retrieved by MOPITT, *Geophys. Res. Lett.*, *31*, L15112, doi:10.1029/2004GL020235.

Deeter, M. N., D. P. Edwards, J. C. Gille, and J. R. Drummond (2007), Sensitivity of MOPITT observations to carbon monoxide in the lower troposphere, *J. Geophys. Res.*, *112*, D24306, doi:10.1029/2007JD008929.

Drummond, J. R., and G. S. Mand (1996), The Measurements of Pollution in the Troposphere (MOPITT) instrument: Overall performance and calibration requirements, *J. Atmos. Oceanic Technol.*, *13*, 314–320.

Edwards, D. P., et al. (2004), Observations of carbon monoxide and aerosols from the Terra satellite: Northern Hemisphere variability, *J. Geophys. Res.*, *109*, D24202, doi:10.1029/2004JD004727.

Edwards, D. P., et al. (2006a), Satellite-observed pollution from Southern Hemisphere biomass burning, *J. Geophys. Res.*, *111*, D14312, doi:10.1029/2005JD006655.

Edwards, D. P., G. Pétron, P. C. Novelli, L. K. Emmons, J. C. Gille, and J. R. Drummond (2006b), Southern Hemisphere carbon monoxide interannual variability observed by Terra/Measurement of Pollution in the Troposphere (MOPITT), *J. Geophys. Res.*, *111*, D16303, doi:10.1029/2006JD007079.

Emmons, L. K., et al. (2004), Validation of Measurements of Pollution in the Troposphere (MOPITT) CO retrievals with aircraft in situ profiles, *J. Geophys. Res.*, *109*, D03309, doi:10.1029/2003JD004101.

Frankenberg, C., U. Platt, and T. Wagner (2005), Retrieval of CO from SCIAMACHY onboard ENVISAT: Detection of strongly polluted areas and seasonal patterns in global CO abundances, *Atmos. Chem. Phys.*, *5*, 1639–1644.

Kar, J., et al. (2004), Evidence of vertical transport of carbon monoxide from Measurements of Pollution in the Troposphere (MOPITT), *Geophys. Res. Lett.*, *31*, L23105, doi:10.1029/2004GL021128.

Lamarque, J. F., et al. (2003), Identification of CO plumes from MOPITT data: Application to the August 2000 Idaho Montana forest fires, *Geophys. Res. Lett.*, *30*(13), 1688, doi:10.1029/2003GL017503.

Lin, Y., C. Zhao, L. Peng, and Y. Fang (2007), Monthly CO emissions calculated using MOPITT and MOZART, *Chin. Sci. Bull.*, *52*(18), 2551–2558.

Luo, M., et al. (2007), Comparison of carbon monoxide measurements by TES and MOPITT: The influence of a priori data and instrument characteristics on nadir atmospheric species retrievals, *J. Geophys. Res.*, *112*, D09303, doi:10.1029/2006JD007663.

Massie, S. T., J. C. Gille, D. P. Edwards, and S. Nandi (2006), Satellite observations of aerosol and CO over Mexico City, *Atmos. Environ.*, *40*, 6019–6031.

McMillan, W. W., C. Barnet, L. Strow, M. T. Chahine, M. L. McCourt, J. X. Warner, P. C. Novelli, S. Korontzi, E. S. Maddy, and S. Datta (2005), Daily global maps of carbon monoxide from NASA's Atmospheric Infrared Sounder, *Geophys. Res. Lett.*, *32*, L11801, doi:10.1029/2004GL021821.

Olivier, J. G. J., J. P. J. Bloos, J. J. M. Berdowski, A. J. H. Visschedijk, and A. F. Bouwman (1999), A 1990 global emission inventory of anthropogenic sources of carbon monoxide on 1 × 1 degree developed in the framework of EDGAR/GEIA, *Chemosphere Global Change Sci.*, *1*, 1–17.

Pétron, G., C. Granier, B. Khattatov, V. Yudin, J.-F. Lamarque, L. Emmons, J. Gille, and D. P. Edwards (2004), Monthly CO surface sources inventory based on the 2000–2001 MOPITT satellite data, *Geophys. Res. Lett.*, *31*, L21107, doi:10.1029/2004GL020560.

Pfister, G., P. G. Hess, L. K. Emmons, J.-F. Lamarque, C. Wiedinmyer, D. P. Edwards, G. Pétron, J. C. Gille, and G. W. Sachse (2005), Quantifying CO emissions from the 2004 Alaskan wildfires using MOPITT CO data, *Geophys. Res. Lett.*, *32*, L11809, doi:10.1029/2005GL022995.

Rodgers, C. D. (2000), *Inverse Methods for Atmospheric Sounding: Theory and Practice, Atmospheric*, *Oceanic and Planet. Phys. Ser.*, vol. 2, World Sci., Hoboken, N. J.

Stavrakou, T., and J.-F. Müller (2006), Grid-based versus big region approach for inverting CO emissions using Measurement of Pollution in the Troposphere (MOPITT) data, *J. Geophys. Res.*, *111*, D15304, doi:10.1029/2005JD006896.

Streets, D. G., et al. (2003), An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, *J. Geophys. Res.*, *108*(D21), 8809, doi:10.1029/2002JD003093.

Tie, X., et al. (2006), Chemical characterization of air pollution in eastern China and the eastern United States, *Atmos. Environ.*, *40*, 2607–2625.

Turquety, S., et al. (2007), Inventory of boreal fire emissions for North America in 2004: The importance of peat burning and pyro-convective injection, *J. Geophys. Res.*, *112*, D12S03, doi:10.1029/2006JD007281.

Turquety, S., et al. (2008), Analysis of CO transport from Asia using nadir (MOPITT, SCIAMACHY) and solar occultation (ACE-FTS) satellite measurements with global modelling, *Atmos. Chem. Phys. Discuss*, *8*, 1–47.

Wang, Y., M. B. McElroy, K. F. Boersma, H. J. Eskes, and J. P. Veefkind (2007), Traffic restrictions associated with the Sino-African summit: Reductions of NO_x detected from space, *Geophys. Res. Lett.*, *34*, L08814, doi:10.1029/2007GL029326.

Yudin, V. A., G. Pétron, J.-F. Lamarque, B. V. Khattatov, P. G. Hess, L. V. Lyjak, J. C. Gille, D. P. Edwards, M. N. Deeter, and L. K. Emmons (2004), Assimilation of the 2000–2001 CO MOPITT retrievals with optimized surface emissions, *Geophys. Res. Lett.*, *31*, L20105, doi:10.1029/2004GL021037.

C. Clerbaux, UPMC Université Paris 6, BP102, Service d'Aéronomie/IPSL, 4 Place Jussieu,75252, Paris Cedex 05, France. (ccl@aero.jussieu.fr)

M. Deeter, D. P. Edwards, L. Emmons, J. Gille, J.-F. Lamarque, S. T. Massie, and X. X. Tie, Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, Colorado, USA.